

**AFOSR-Taiwan Nanoscience Initiative**

**Project Final Report**

**Project Title**

**GaN/AlGaN Terahertz Quantum Cascade Laser**  
**FA5209-04-T-0349**

**Period : September 1, 2004- August 31, 2005**

**Principal Investigator**

**Professor S. C. Wang**

**Institute**

Institute of Electro-Optical Engineering,  
National Chiao Tung University  
1001 Ta Hsueh Road, Hsinchu, TAIWAN 30010  
Phone: +886-3-5712121 extension 56320  
Fax: +886-3-5716631  
Email: [scwang@mail.nctu.edu.tw](mailto:scwang@mail.nctu.edu.tw)

**October 7, 2005**

Report Documentation Page				Form Approved OMB No. 0704-0188	
Public reporting burden for the collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to a penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.					
1. REPORT DATE <b>16 FEB 2007</b>		2. REPORT TYPE <b>FInal</b>		3. DATES COVERED <b>21-07-2004 to 20-12-2005</b>	
4. TITLE AND SUBTITLE <b>GaN/AlGaIn Terahertz Quantum Cascade Laser</b>				5a. CONTRACT NUMBER <b>FA520904P0458</b>	
				5b. GRANT NUMBER	
				5c. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S) <b>Shing-Chung Wang</b>				5d. PROJECT NUMBER	
				5e. TASK NUMBER	
				5f. WORK UNIT NUMBER	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) <b>National Chiao Tung University,1001 Ta Hsueh Rd,Hsinchu 300,Taiwan,TW,300</b>				8. PERFORMING ORGANIZATION REPORT NUMBER <b>N/A</b>	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) <b>AOARD, UNIT 45002, APO, AP, 96337-5002</b>				10. SPONSOR/MONITOR'S ACRONYM(S) <b>AOARD</b>	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) <b>AOARD-044070</b>	
12. DISTRIBUTION/AVAILABILITY STATEMENT <b>Approved for public release; distribution unlimited</b>					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT <b>The quantum-cascade laser (QCL) has, since its first realization, demonstrated an impressive and rapid development, extending the emission wavelengths from midinfrared to terahertz spectral range. However, QCLs based on GaAs/AlGaAs and AlInAs/GaInAs are not capable of emitting in the energy range around the LO-phonon energies (ELO~36 meV in GaAs and ELO~34 meV in InGaAs), leaving a gap in the spectral scale between 30 and 40 &amp;#956;m. This can be overcome by using GaN/AlGaIn material. The GaN based QCL has many advantages over the GaAs QCL. These include larger LO-phonon energy (ELO~90 meV), very fast carrier dynamics, far infrared emission wavelengths (&gt; 40 &amp;#956;m), and room temperature operation capability. Therefore it has been considered the most desirable candidate for far infrared intersubband emission laser.</b>					
15. SUBJECT TERMS <b>Nanotechnology</b>					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT <b>Same as Report (SAR)</b>	18. NUMBER OF PAGES <b>11</b>	19a. NAME OF RESPONSIBLE PERSON
a. REPORT <b>unclassified</b>	b. ABSTRACT <b>unclassified</b>	c. THIS PAGE <b>unclassified</b>			

## 1. Introduction

The quantum-cascade laser (QCL) has, since its first realization, demonstrated an impressive and rapid development, extending the emission wavelengths from mid-infrared to terahertz spectral range. However, QCLs based on GaAs/AlGaAs and AlInAs/GaInAs are not capable of emitting in the energy range around the LO-phonon energies ( $E_{LO} \sim 36$  meV in GaAs and  $E_{LO} \sim 34$  meV in InGaAs), leaving a gap in the spectral scale between 30 and 40  $\mu\text{m}$ . This can be overcome by using GaN/AlGaN material. The GaN based QCL has many advantages over the GaAs QCL. These include larger LO-phonon energy ( $E_{LO} \sim 90$  meV), very fast carrier dynamics, far infrared emission wavelengths ( $> 40$   $\mu\text{m}$ ), and room temperature operation capability. Therefore it has been considered the most desirable candidate for far infrared intersubband emission laser. Recent analysis by Prof. Greg Sun and Dr. Richard Soref suggested that GaN based QCL can be realized using the GaN/AlGaN cascade structure as shown in Fig. 1. Our research group has been conducting research on GaN based optoelectronic materials and light emitting devices for past several years using the MOCVD system for epitaxial growth of GaN based materials and structures. We have grown various GaN-based light emitting devices including LEDs and lasers and quantum confined structures under the sponsorship of our National Science Council (NSC). In this initial one-year program with AFOSR, our effort has been focused on the MOCVD growth of the GaN/AlGaN quantum confined structures related to the proposed GaN QCL design shown in Fig.1. During this period we have established the growth conditions for obtaining good surface morphology of the epitaxial structures and the growth of active layer structure. In particular we have grown and demonstrated a 60-period high-quality AlGaN/GaN MQW structure similar to the one proposed in Fig. 1. We also conducted investigation and analysis of the grown structure to establish

the compositional contents and thickness of the grown active layer structures which are very important parameters for growth of the overall GaN QCL structure. We also presented one paper in collaboration with Prof. Greg Sun and Dr. Richard Soref at the US Air Force/Taiwan Nanoscience Initiative Workshop in February 2005 in Hawaii, and submitted a paper to Applied Physics Letters in August 2005. In this final report, we summarize our investigation results and accomplishments in these key areas.

## 2. Results and Accomplishments

### 2.1 Growth and characterization of single AlGa<sub>x</sub>N epilayer

The surface morphology and the exact compositional contents of the GaN/Al<sub>x</sub>Ga<sub>1-x</sub>N epitaxial structure are very important factors in the realization of the GaN/AlGa<sub>x</sub>N QCL structure. We have grown the AlGa<sub>x</sub>N epilayers on sapphire to investigate their morphologies and aluminum compositions. The surface image of the grown Al<sub>0.2</sub>Ga<sub>0.8</sub>N sample observed by AFM is shown in Fig.2. The surface is very smooth and the line profile also shows crack free. The surface roughness of this sample is about 0.24 nm. Fig.3 shows the double crystal x-ray diffraction rocking curve (0004) of Al<sub>x</sub>Ga<sub>1-x</sub>N samples. The Al composition can be calculated from the diffraction angle separation between AlGa<sub>x</sub>N and GaN peaks. The room temperature photoluminescence spectra of Al<sub>x</sub>Ga<sub>1-x</sub>N films are shown in Fig. 3. The nonlinear dependence of the energy gap on Al composition can then be described by

$$E_g^{Al_xGa_{1-x}N}(x) = E_g^{GaN}(1-x) + E_g^{AlN}x - bx(1-x) \quad (1)$$

Where  $b$  is the direct gap bowing parameter, and  $E_g^{AlN}$  were determined to be 6.13 eV at room temperature. For the determination of  $E_g^{GaN}$ , the peak energy of PL is 3.41. We further take into account that PL peak position is 0.03 eV lower than the gap energy determined by absorption measurements. For the evaluation of the Al<sub>x</sub>Ga<sub>1-x</sub>N data, a

value of  $E_g^{GaN}=3.44$  eV was chosen. In Fig. 5 the alloy band gap is shown as a function of the chemical composition which was determined by XRD. The bowing parameters can be fitted with  $b = 0.86$  eV and  $b = 1.3$  eV. Beside five points fitted with  $b = 0.86$  eV, one can notice most of them are fitted with  $b = 1.3$  eV. This result is not only important for future growth of the QCL structure but also is an important data for  $Al_xGa_{1-x}N$  materials. We will submit a paper on this important result for publication soon.

## 2.2 Growth and investigation of active region of quantum cascade laser

We have grown a multilayer structure of active region of GaN/AlGa<sub>N</sub> MQW structure which consists of 3 GaN QWs and 3 AlGa<sub>N</sub> barriers as shown in Fig. 6. The AlGa<sub>N</sub>/GaN MQW structures were grown on (0001) sapphire substrates using low-pressure VEECO D75 MOCVD system (original EMCORE MOCVD system). Trimethylgallium (TMG), TMA, and NH<sub>3</sub> were used as source materials with H<sub>2</sub> and N<sub>2</sub> carrier gases. The 60-period AlGa<sub>N</sub>/GaN MQW structures were realized by 20 time repetitions of the growth of a 3-period MQW and a 2-μm-thick GaN buffer layer in order to suppress a formation of cracks in the MQWs. Surface morphology was examined using an optical microscope. The crystalline quality of the samples was evaluated by (0002) symmetric high-resolution x-ray diffraction (HRXRD) using four-crystal (022) Ge monochromators with a Cu  $K_{\alpha 1}$  radiation. The average thicknesses of the AlGa<sub>N</sub> barriers and the GaN wells were determined by the angular distance using satellite peaks of  $\omega/2\theta$ -scan diffraction patterns. A transmission electron microscope (TEM) was used to study thickness and the sharpness of the QC structure interfaces.

Figure 7 showed very smooth surface morphology for all the samples, but a few cracks were also observed at the edges of the 2-in. wafers. We therefore consider that the GaN buffer layers play a role to suppress the generation of cracks as expected. Figure 8 shows (a) a cross-sectional TEM image of the 20-period QC structure and

each period consisting of 3 GaN QWs and 3 AlGaN barriers and (b) two periods of the QC structure. In Fig. 8(a), a contrast made by the periodically aligned 20 sets of the 3-period MQWs can be seen. No dislocations running across the sample are seen in this figure. In Fig. 8(b), bright parts and dark parts correspond to the AlGaN barriers and the GaN wells, respectively. It is obviously seen that the interfaces are very sharp. Note that the interfaces of the AlGaN barriers on the GaN wells are as sharp as those of the GaN wells on the AlGaN barriers. From this figure, the error of the thickness of the AlGaN barriers and GaN wells compare to design can be estimated to be 0.5  $\mu\text{m}$ .

The HRXRD pattern of the  $\omega/2\theta$ -scan (0002) reflections for QC structure is shown in Fig. 9. The solid and dashed lines show the experimental and simulated results, respectively. The satellite peaks due to the QC structure can be clearly seen up to the 4<sup>th</sup> order peaks indicates that smooth and abrupt interfaces with good periodicity of the QC structure, even through 20 cascading periods have been stacked. Table 1 has listed the thicknesses and compositions of epilayers in one period. The good agreement between the experimental curve and the simulated data confirm the successful fabrication of a QC structure, but there are 0.5 nm error in thicknesses. The aluminum composition of multiple quantum well is different from the single AlGaN epilayer even if the gas composition  $\text{TMAI}/(\text{TMGa}+\text{TMGa})$  kept constant in the growth.

In summary, we have grown high crystalline quality AlGaN single layer and active region of QCL by low-pressure MOCVD. The morphologies of single AlGaN and full active region are quite smooth. The compositional data of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  were determined and the initial active layer structure of QCL was successfully grown and investigated. Further research on the precision controls of thickness and fine tuning of the composition are needed in the next phase of the program in order to grow the full QCL structure and realization of the GaN/AlGaN QCL.

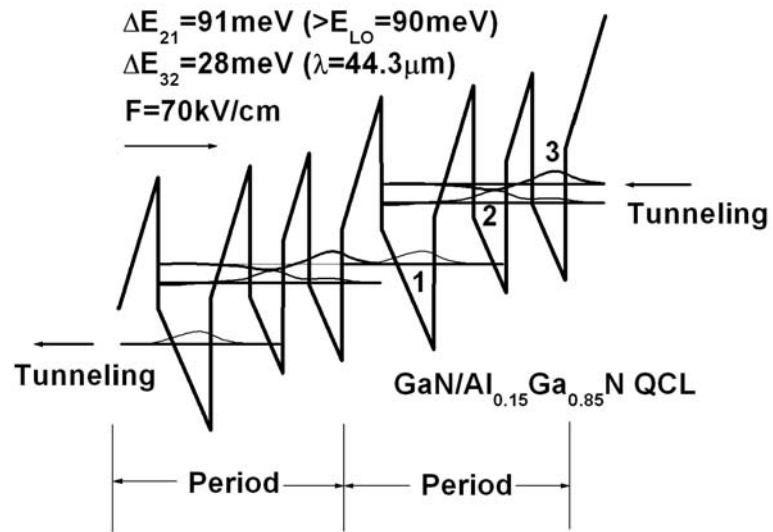


Fig. 1 Proposed GaN/AlGa<sub>0.85</sub>N QCL structure

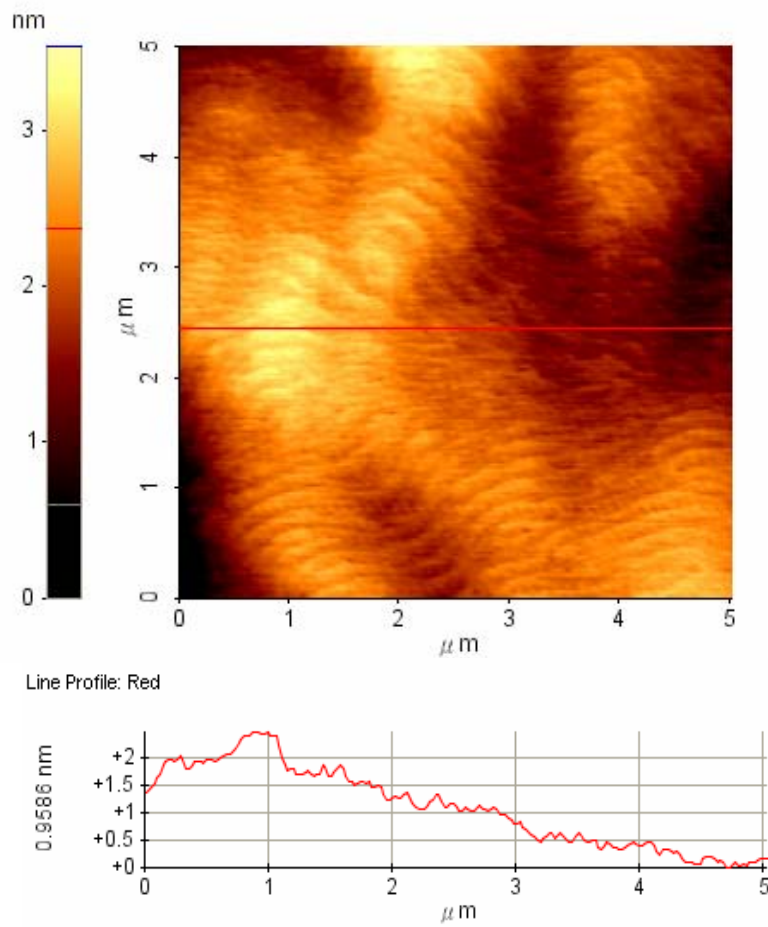


Fig. 2 The AFM surface image of Al<sub>0.2</sub>Ga<sub>0.8</sub>N grown sample.

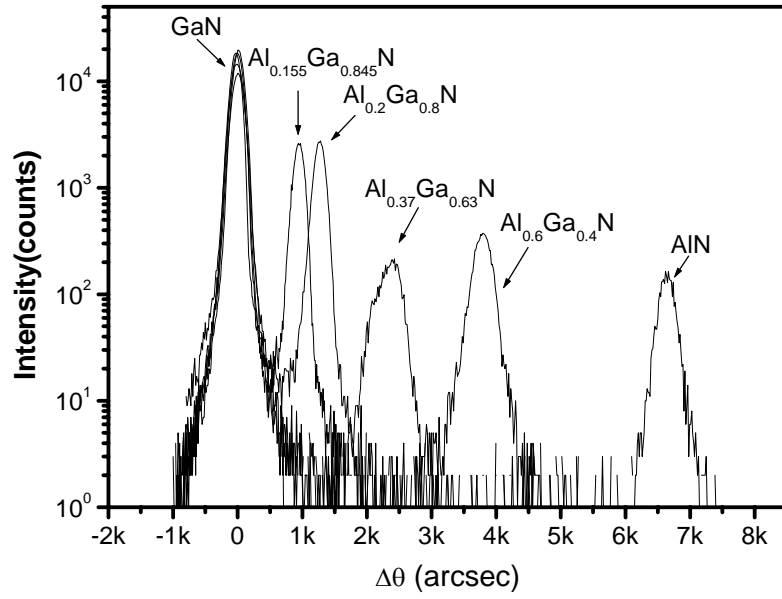


Fig. 3 x-ray double crystal rocking curve (0004) of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  samples grown on GaN/sapphire substrate.

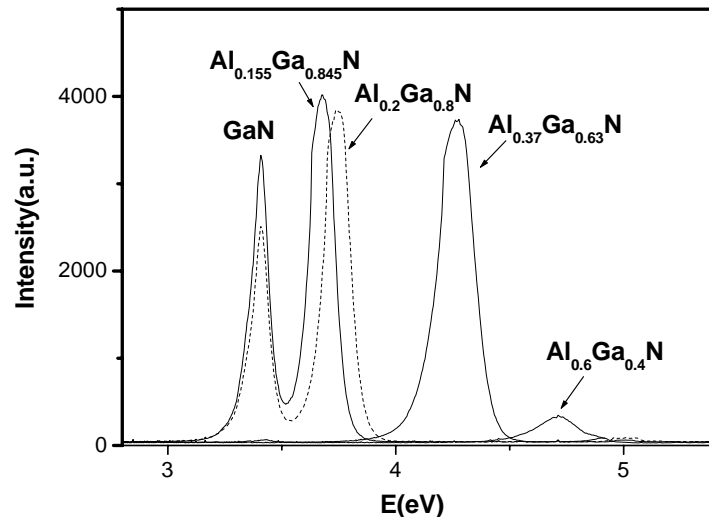


Fig. 4 Room temperature PL spectra of  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  samples with different Al compositions.



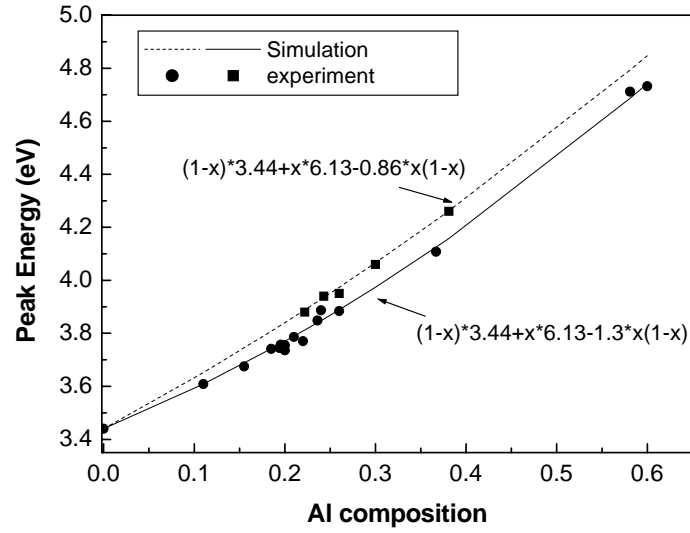


Fig. 5 Band gap of the epitaxial  $\text{Al}_x\text{Ga}_{1-x}\text{N}$  films vs Al composition. The solid line was calculated used Eq. 1 and bowing parameter of 0.86 eV and 1.3 eV, respectively.

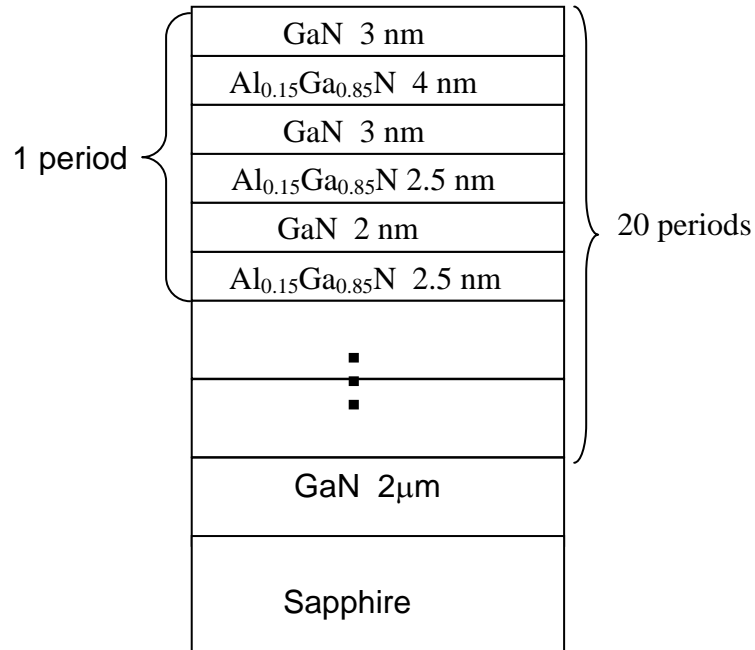


Fig. 6 Schematic of AlGaIn QCL Active Layer Structure

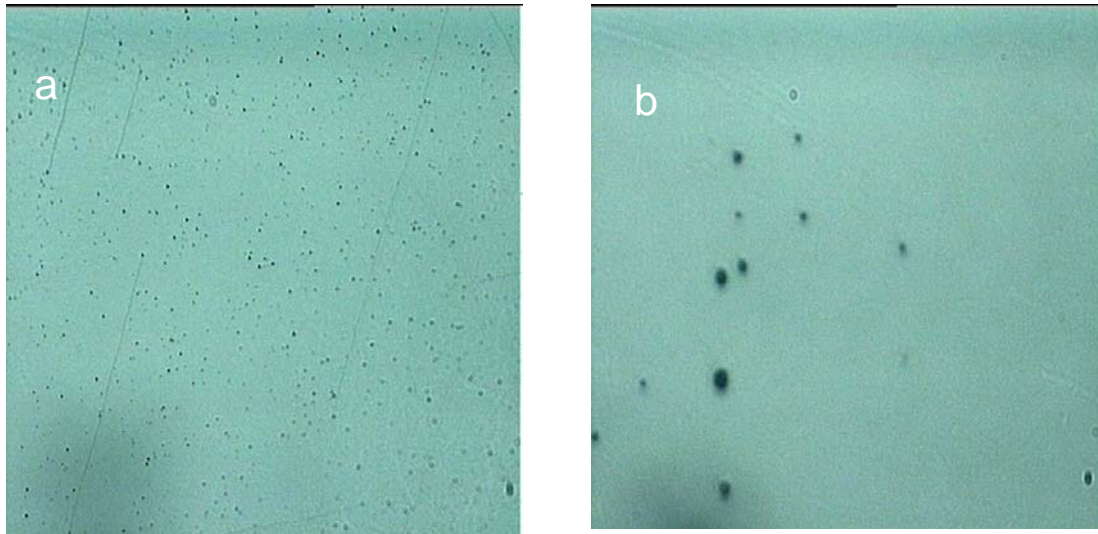


Fig.7 Microscopy surface image of AlGaIn/GaN QCL structure (a)  $\times 10$  (b)  $\times 100$

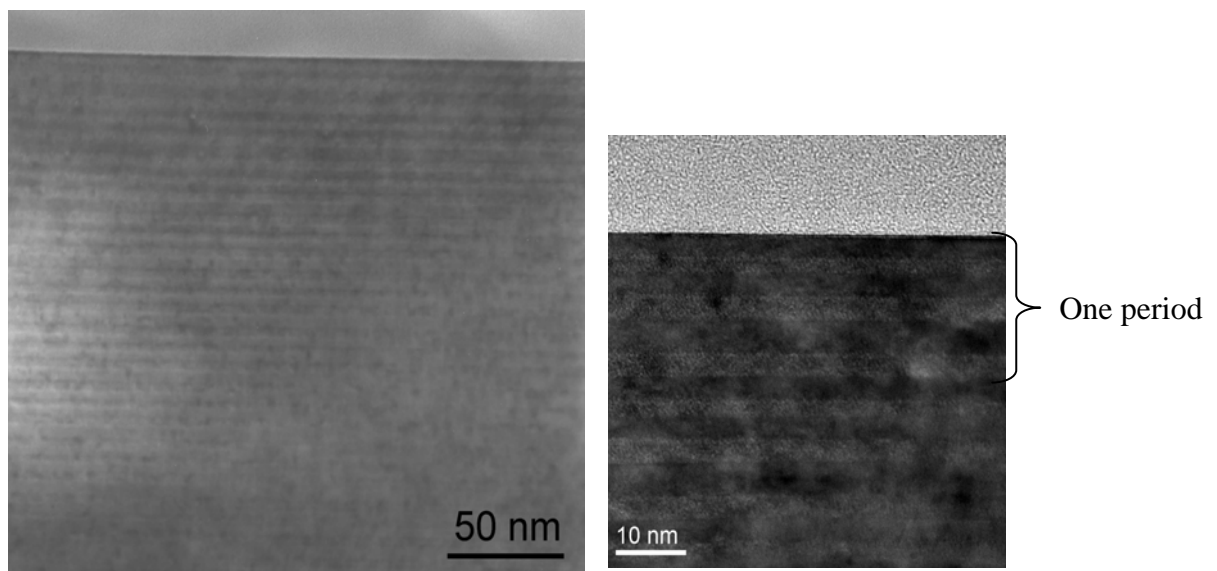


Fig. 8 Transmission electron microscope image of (a) the portion of the QCL structure and (b) two periods of QWs. Light grey layers indicate  $\text{Al}_{0.15}\text{Ga}_{0.85}\text{N}$  and dark grey layers GaN.

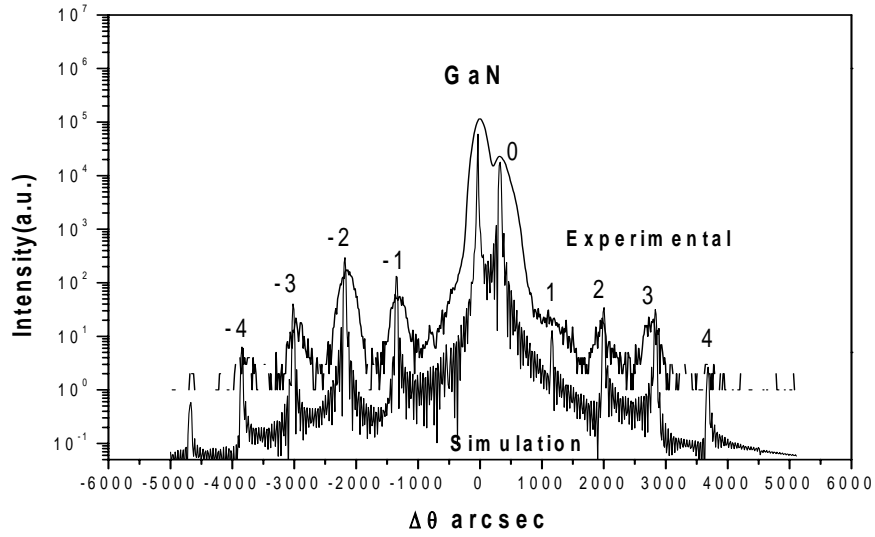


Fig.9 (0 0 0 2)  $\omega$ -2 $\theta$  x-ray diffraction pattern and simulated result of AlGaIn/GaN quantum cascade laser active region structure.

Table 1. Simulation Results of x-ray diffraction pattern

layer	Thickness(Å)	Material	Al composition
1	29	GaN	
2	24	Al <sub>x</sub> Ga <sub>1-x</sub> N	0.20
3	29	GaN	
4	36	Al <sub>x</sub> Ga <sub>1-x</sub> N	0.20
5	45	GaN	
6	36	Al <sub>x</sub> Ga <sub>1-x</sub> N	0.20

## References

1. G.S. Huang, H. H. Yao, H.C. Kuo, T. C. Lu, S. C. Wang, Chih-wei Lin and Li Chang, "Crack-free GaN/AlN distributed Bragg reflectors grown on sapphire using GaN/AlN superlattices for strain management" submitted to Applied Physics Letter.
2. Greg Sun, Richard A. Soref, Jacob B. Khurgin, "Active region design of a terahertz GaN/Al<sub>0.15</sub>Ga<sub>0.85</sub>N quantum cascade laser," to be published in *Superlattices and Microstructures*, 2004
3. H. H. Yao, C. F. Lin, H. C. Kuo, S. C. Wang, "MOCVD growth of AlN/GaN DBR structures under various ambient conditions," J. Crystal Growth 262 (2004), 151-156
4. C. C. Kao, H. W. Huang, J. Y. Tsai, C. C. Yu, C. F. Lin, H. C. Kao, S. C. Wang, "Study of dry etching for GaN and InGaN-based laser structure using inductively coupled plasma reactive ion etching," Materials Sciences and Engineering B107(2004), 283-288.
5. F. Yun, M. A. Reshchikov, L. He, T. King, H. Morkoc, S. W. Novak, L. Wei, J. Appl. Phys. 92(2002) 4837.